

DECLARATION

DEC 0 9 2003 TC 1700

I, Shigeaki Nishikawa of Nishikawa & Associates, Visual City, Suite 401, 43-9, Higashi-Nippori 3-chome, Arakawa-ku, Tokyo, Japan, do solemnly and sincerely declare that I am well acquainted with both the Japanese language and the English language and that the attached English translation of an officially certified copy of Patent Application No. 122999/1999 is a true and correct translation to the best of my knowledge and belief from the Japanese language to the English language.

Dated this 1st day of December, 2003

Shigeaki Nishikawa

Suzare Klishi Doch

(translator)

(Translation)

PATENT OFFICE JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of the following application as filed with this Office.

Date of Application: April 28, 1999

Application Number: Patent Application No. 122999/1999

Applicant(s): NIPPON ZEON CO., LTD.

Dated: June 2, 2000

Takahiko Kondo Commissioner, Patent Office

Certificate No. 2000-3040954

[Document Name] Application for Patent [Docket Number] PN990036 [To] Hon. Director General of Patent Office [International Patent Classification] 5 B29C [Inventor] [Address or Domicile] c/o Research & Development Center, Nippon Zeon Co., Ltd., 2-1, Yako 1-chome Kawasaki-ku, Kawasaki-shi, Kanagawa 10 [Name] Keiichi Kawata [Inventor] [Address or Domicile] c/o Research & Development Center, Nippon Zeon Co., Ltd., 2-1, Yako 1-chome Kawasaki-ku Kawasaki-shi, Kanagawa 15 [Name] Naoki Shinohara [Applicant for Patent] [Identification Number] 000229117 [Name] NIPPON ZEON CO., LTD. [Representative] Katsuhiko Nakano 20 [Indication of Fee] [Deposit Account Number] 033684 [Fee] ¥21,000 [List of Materials Submitted] [Material Name] Specification

Abstract

1

25

[Material Name]

[Document Name] Specification

[Title of the Invention] LAMINATE AND PRODUCTION PROCESS THEREOF

[Claims]

[Claim 1] A laminate comprising at least one alicyclic polymer layer, at least one layer containing an alicyclic polymer and a thermoplastic resin, and at least one thermoplastic resin layer.

[Claim 2] A process for producing a laminate, which
comprises melting and kneading a laminate material
comprising at least one alicyclic polymer layer and at
least one thermoplastic resin layer, and then co-extruding
the kneaded product and other polymers or resins.
[Detailed Description of the Invention]

15 [0001]

20

[Technical Field to which the Invention Belongs]

The present invention relates to a laminate and a production process thereof, and particularly to a laminate which is free of clouding, high in transparency and heat resistance, excellent in tearability or cuttability, good in adhesion to others, and a production process thereof. [0002]

[Prior Art]

Many laminates such as laminate made by vinyl

25 chloride resins have heretofore been used as a laminate in
food packaging films, agricultural films, drug packaging
films, containers, etc. However, laminates containing

materials possibly produce substances such as dioxin and environmental hormones, which exert an adverse influence on life environment and living bodies, have been going to be changed to resin laminates having high environmental safety such as polyethylene, and many researches and developments have come to be made.

5

10

In the course of such researches and developments, there have been proposed single-layer films composed of a alicyclic polymer such as a norbornene addition polymer and laminated films obtained by laminating a polyethylene layer and a norbornene addition polymer layer.

The single-layer films composed of the norbornene addition polymer are high in transparency and low in hygroscopicity. Since such a single-layer film is insufficient in oil resistance or resistance to greases and oils, however, the film tends to cause cracking and clouding when it is used in such a form that it comes into direct contact with food.

Further, a laminate obtained by laminating a

20 norbornene addition polymer and a layer composed of
polyolefin or polyamide has been easy to cause such
inconvenience that when it is applied to use applications,
in which the laminated film is stretched, white wrinkles
occur at a stretched portion, or one layer is broken due to

25 delamination because the elongation percentages of the
individual layer are different and the compatibility of the
norbornene addition polymer with polyolefin or polyamide is

insufficient.

.:

A film formed by a composition comprising a polyolefin and a norbornene addition polymer has been proposed, however, it is difficult to provide a film having high transparency and excellent surface smoothness while retaining the desired strength because of the low compatibility of the polyolefin and the norbornene addition polymer.

[0003]

15

20

25

10 [Problems Sought for Solution by the Invention]

It is an object of the present invention to provide a laminate which contains a layer containing a polyolefin and an alicyclic polymer and is free of clouding, high in transparency and heat resistance, excellent in smoothness, tearability or cuttability, low moisture permeability and good in adhesion to others equal to or higher than those properties of a norbornene single layer or a laminated film obtained by laminating a norbornene polymer and a polyolefin, more specifically, to provide a laminated film having such properties.

Another object of the present invention is to provide a production process of a laminate by effectively utilizing waste materials discharged in the production step of the laminate to obtain the laminate which has above-described properties.

[0004]

The present inventors have carried out an extensive

investigation with a view toward achieving the above objects. As a result, it has been found that when an alicyclic polymer layer, a layer containing an alicyclic polymer and a thermoplastic resin, and a thermoplastic resin layer are laminated, a laminate can be provided as a laminate free of clouding, high in transparency and heat resistance, excellent in surface smoothness and tearability or cuttability and good in adhesion to others. It has been also found that the above-described laminate can be preferably produced by melting and kneading a laminate 10 comprising at least one alicyclic polymer layer and at least one thermoplastic resin layer, and then co-extruding the kneaded product with other polymers or resins. present invention has been led to completion on the basis 15 of these findings.

[0005]

20

[Means for solution of the Problems]

According to the present invention, there is thus provided a laminate comprising at least one alicyclic polymer layer, at least one layer containing an alicyclic polymer and a thermoplastic resin, and at least one thermoplastic resin layer.

According to the present invention, there is also provided a process for producing a laminate, which

comprises melting and kneading a laminate material comprising at least one alicyclic polymer layer and at least one thermoplastic resin layer, and then co-extruding

the kneaded product and other polymers or resins. [0006]

[Mode for Carrying out the Invention]

The laminates according to the present invention have

5 a structure that at least one alicyclic polymer layer, at
least one layer containing an alicyclic polymer and a
thermoplastic resin, and at least one thermoplastic resin
layer are laminated.

[0007]

(خ

10 The alicyclic polymer used in the present invention is a polymer having an alicyclic structure at its main chain and/or side chain. The alicyclic polymer preferably has the alicyclic structure at its main chain from the viewpoints of mechanical strength, heat resistance and the like. Examples of the alicyclic structure include a 15 cycloalkane structure, a cycloalkene structure and the like. The cycloalkane structure is preferred from the viewpoints of mechanical strength, heat resistance and the like. Examples of the alicyclic structure include a monocycle, polycycle, a fused polycycle, a crosslinked cycle and 20 combined polycycles thereof. No particular limitation is imposed on the number of carbon atoms forming the alicyclic structure. However, it is within a range of generally 4 to 30 carbons, preferably 5 to 20 carbons, more preferably 5 to 15 carbons. Various properties such as mechanical 25 strength, heat resistance and moldability are balanced with

one another at a high level by such an alicyclic structure.

[8000]

· 3

A proportion of the repeating unit having the alicyclic structure in the alicyclic structure polymer can be suitably selected as necessary for the end application intended. However, it is generally at least 30% by weight, 5 preferably at least 50% by weight, more preferably at least 70% by weight. The upper limit of the proportion is 100% by weight. If the proportion of the repeating unit having the alicyclic structure in the alicyclic polymer is too low, 10 the heat resistance of the alicyclic polymer becomes poor. It is hence not preferable to contain such a repeating unit in a too low proportion. No particular limitation is imposed on other repeating units than the repeating unit having the alicyclic structure in the alicyclic polymer, and they are suitably selected as necessary for the end 15 application intended. More specifically, as the alicyclic polymer, may be used not only a homopolymer of a monomer having an alicyclic structure or a copolymer of such alicyclic monomers, but also a copolymer of the alicyclic monomer with a non-alicyclic monomer copolymerizable 20 therewith. The alicyclic polymer may be subjected to such a treatment that an unsaturated bond is hydrogenated into a saturated bond.

[0009]

As examples of the alicyclic polymer, may be mentioned norbornene polymers, monocyclic cycloolefin polymers, cyclic conjugated diene polymers, vinyl cyclic

hydrocarbon polymers, and hydrogenated products thereof.

Among these, the norbornene polymers and hydrogenated products thereof, and the cyclic conjugated diene polymers and hydrogenated products thereof are preferred.

5 [0010]

No particular limitation is imposed on the norbornene polymers, and examples thereof include polymers obtained by subjecting a norbornene monomer to polymerization in accordance with, for example, the process disclosed in 10 Japanese Patent Application Laid-Open No. 14882/1991 or 122137/1991. Specific examples of the norbornene polymers include ring-opening polymers of norbornene monomer(s) and hydrogenated products thereof, addition polymers of norbornene monomer(s) and addition polymers of a norbornene monomer and a vinyl monomer. Of these, the hydrogenated 15 products of the ring-opening polymers of the norbornene monomer(s), addition polymers of norbornene monomer(s) and addition polymers of a norbornene monomer and a vinyl monomer copolymerizable therewith are particularly preferred from the viewpoints of good balance between heat 20 resistance, dielectric constant, and the like.

[0011]

The norbornene monomers are publicly known monomers disclosed in the above-described publications, Japanese

25 Patent Application Laid-Open Nos. 227424/1990 and 276842/1990, etc. Examples of the norbornene monomers include bicyclo[2.2.1]hept-2-ene (trivial name: norbornene),

5- methyl-bicyclo[2.2.1]hept-2-ene, 5,5-dimethyl-bicyclo[2.2.1]hept-2-ene, 5-ethyl-bicyclo[2.2.1]hept-2-ene, 5butyl-bicyclo[2.2.1]hept-2-ene, 5-hexyl-bicyclo[2.2.1]hept2-ene, 5-octyl-bicyclo[2.2.1]hept-2-ene, 5-octadecylbicyclo[2.2.1]hept-2-ene, 5-ethylidene-bicyclo[2.2.1]hept2-ene, 5-methylidene-bicyclo[2.2.1]hept-2-ene, 5-vinylbicyclo[2.2.1]hept-2-ene,

[0012]

· 1 . 🖎

5-propenyl-bicyclo[2.2.1]hept-2-ene, 5
methoxycarbonyl-bicyclo[2.2.1]hept-2-ene, 5-cyanobicyclo[2.2.1]hept-2-ene, 5-methyl-5-methoxycarbonylbicyclo[2.2.1]hept-2-ene, 5-ethoxycarbonyl-bicyclo[2.2.1]hept-2-ene, bicyclo[2.2.1]hept-5-enyl 2-methylpropionate, bicyclo[2.2.1]hept-5-enyl 2-methyloctanate,

[0013]

Bicyclo[2.2.1]hept-2-ene-5,6-dicarboxylic acid
anhydride, 5-hydroxymethylbicyclo[2.2.1]hept-2-ene, 5,6di(hydroxy- methyl)-bicyclo[2.2.1]hept-2-ene, 5hydroxyisopropyl-bicyclo[2.2.1]hept-2-ene, 5,6-dicarboxybicyclo[2.2.1]hept-2-ene, bicyclo[2.2.1]hept-2-ene-5,6dicarboxylic acid imide, 5-cyclopentyl-bicyclo[2.2.1]hept2-ene, 5-cyclohexyl-bicyclo[2.2.1]hept-2-ene, 5cyclohexenyl-bicyclo-[2.2.1]hept-2-ene, 5-phenylbicyclo[2.2.1]hept-2-ene,

25 [0014]

Tricyclo[$4.3.0.1^{2.5}$]dec-3.7-diene (trivial name: dicyclopentadiene), tricyclo[$4.3.0.1^{2.5}$]dec-3-ene,

tricyclo[$4.4.0.1^{2.5}$]undec-3,7-diene, tricyclo[$4.4.0.1^{2.5}$]-undec-3,8-diene, tricyclo[$4.4.0.1^{2.5}$]undec-3-ene, tetracyclo[$7.4.0.1^{10.13}.0^{2.7}$]tridec-2,4,6,11-tetraene (another name: 1,4-methano-1,4,4a,9a-tetrahydrofluorene), tetracyclo[$8.4.0.1^{11.14}.0^{3.8}$]tetradec-3,5,7,12-11-tetraene (another name: 1,4-methano-1,4,4a,5,10,10a-hexahydro-anthracene),

[0015]

Tetracyclo $[4.4.0.1^{2.5}.1^{7.10}]$ -dodec-3-ene (trivial name:

- 10 tetracyclododecene), 8-methyltetracyclo-
 - $[4.4.0.1^{2.5}.1^{7.10}]$ dodec-3-ene, 8-ethyltetracyclo-
 - $[4.4.0.1^{2.5}.1^{7,10}]$ dodec-3-ene, 8-methylidenetetracyclo-
 - $[4.4.0.1^{2.5}.1^{7.10}]$ dodec-3-ene, 8-ethylidenetetracyclo-
 - $[4.4.0.1^{2.5}.1^{7.10}]$ dodec-3-ene, 8-vinyltetracyclo-
- 15 $[4.4.0.1^{2.5}.1^{7.10}]$ dodec-3-ene, 8-propenyltetracyclo-
 - $[4.4.0.1^{2.5}.1^{7.10}]$ dodec-3-ene, 8-methoxycarboxy-tetracyclo-
 - $[4.4.0.1^{2.5}.1^{7.10}]$ dodec-3-ene, 8-methyl-8-methoxycarbonyl-

 $\texttt{tetracyclo[4.4.0.1}^{2,5}.1^{7,10}] \, \texttt{dodec-3-ene, 8-hydroxymethyl-}$

 $tetracyclo[4.4.0.1^{2.5}.1^{7.10}]dodec-3-ene, 8-carboxy-$

20 tetracyclo[$4.4.0.1^{2,5}.1^{7,10}$] dodec-3-ene,

[0016]

8-cyclopentyl-tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8-cyclohexyl-tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8cyclohexenyl-tetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, 8phenyl-tetracyclo-[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene, pentacyclo[6.5.1.1^{3,6}.0^{2,7}.0^{9,13}]-pentadec-3,10-diene and pentacyclo[7.4.0.1^{3,6}.1^{10,13}.0^{2,7}]-pentadec-4,11-diene. [0017]

. . . .

5

10

These norbornene monomers may be used either singly or in any combination thereof. The norbornene polymer may be a copolymer of a norbornene monomer and another monomer copolymerizable therewith. The content of bound norbornene monomer units in the norbornene polymer is suitably selected as necessary for the end application intended. However, it is generally at least 30% by weight, preferably at least 50% by weight, more preferably at least 70% by weight, whereby the dielectric constant, the heat resistance and elongation properties of such a polymer can be balanced with each other at a high level.

[0018]

Examples of the vinyl monomer copolymerizable with the norbornene monomer include ethylenes or $\alpha\text{-olefins}$ 15 having 2 to 20 carbon atoms, such as ethylene, propylene, 1-butene, 1-pentene, 1-hexene, 3-methyl-1-butene, 3-methyl-1-pentene, 3-ethyl-1-pentene, 4-methyl-1-pentene, 4-methyl-1-hexene, 4,4-dimethyl-1-hexene, 4,4-dimethyl-1-pentene, 4ethyl-1-hexene, 3-ethyl-1-hexene, 1-octene, 1-decene, 1-20 dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene and 1eicosene; cycloolefins such as cyclobutene, cyclopentene, cyclohexene, 3,4-dimethylcyclopentene, 3-methylcyclohexene, 2-(2-methylbutyl)-1-cyclohexene, cyclooctene and 3a,5,6,7atetrahydro-4,7-methano-1H-indene; and non-conjugated dienes 25 such as 1,4-hexadiene, 4-methyl-1,4-hexadiene, 5-methyl-1,4-hexadiene and 1,7-octadiene. These vinyl monomers may

be used either singly or in any combination thereof. [0019]

No particular limitation is imposed on the polymerization process of the norbornene monomer, or the norbornene monomer and the vinyl monomer copolymerizable with the norbornene monomer, and a hydrogenation process. The polymerization and hydrogenation may be conducted in accordance with any publicly-known processes.

[0020]

As examples of the monocyclic cycloolefin polymer, may be mentioned addition (co)polymers of a monocyclic cycloolefin monomer such as cyclohexene, cycloheptene or cyclooctene, which are disclosed in Japanese Patent Application Laid-Open No. 66216/1989.

15 [0021]

20

. : . .

5

As examples of the cyclic conjugated diene polymer, may be mentioned (co)polymers obtained by subjecting a cyclic conjugated diene such as cyclopentadiene or cyclohexadiene to 1,2- or 1,4-addition polymerization, and hydrogenated products thereof, which are disclosed in Japanese Patent Application Laid-Open Nos. 136057/1994 and 258318/1995.

[0022]

As examples of the vinyl alicyclic hydrocarbon

25 polymer, may be mentioned polymers of a vinyl cyclic
hydrocarbon monomer such as vinylcyclohexene or
vinylcyclohexane, and hydrogenated products thereof, which

are disclosed in Japanese Patent Application Laid-Open No. 59989/1976, and polymers of a vinyl aromatic monomer such as styrene or α -methylstyrene, the aromatic ring portions of which have been hydrogenated, and which are disclosed in Japanese Patent Application Laid-Open Nos. 43910/1988 and 1706/1989.

[0023]

.

5

The alicyclic polymer may have a polar group such as a hydroxyl group or carboxyl group.

- The alicyclic polymer having a polar group is obtained by, for example, (1) introducing a compound having a polar group into said alicyclic polymer by a modification reaction, or (2) copolymerizing a monomer containing a polar group as a comonomer.
- Examples of the polar group include hydroxyl, carboxyl, oxy, epoxy, glycidyl, oxycarbonyl, carbonyloxy, carbonyl and amino groups.

[0024]

The alicyclic polymers may be used either singly or 20 in any combination thereof.

[0025]

No particular limitation is imposed on the molecular weight of the alicyclic polymer. The molecular weight of the alicyclic polymer is generally 1,000 to 1,000,000,

preferably 5,000 to 500,000, more preferably 10,000 to 250,000 when expressed by a weight average molecular weight (Mw) in terms of polystylene as measured by gel permeation

chromatography (GPC) using cyclohexane or toluene as a solvent. It is preferable that the weight average molecular weight (Mw) of the alicyclic polymer falls within this range because the heat resistance and adhesion property of the polymer and the surface smoothness of the resulting laminate are balanced with each other at a high level.

[0026]

5

15

20

25

The molecular weight distribution of the alicyclic

10 polymer is expressed by a ratio (Mw/Mn) of the weight
average molecular weight (Mw) to the number average
molecular weight (Mn) as measured by GPC using cyclohexane
or toluene as a solvent and is generally at most 5,
preferably at most 4, more preferably at most 3.

The ranges of the weight average molecular weight (Mw) and molecular weight distribution (Mw/Mn) and the measuring methods thereof are particularly suitable for the norbornene polymers. However, the present invention is not limited thereto. In the case of an alicyclic polymer the weight average molecular weight and molecular weight distribution of which cannot be measured by the above-described method, that having a melt viscosity or polymerization degree to such an extent that a resin layer can be formed by an ordinary melt processing process may be used.

[0027]

The glass transition temperature of the alicyclic

polymer may be suitably selected as necessary for the end application intended. However, it is generally at least 50°C, preferably at least 70°C, more preferably at least 100°C, most preferably at least 125°C.

5 [0028]

Examples of the thermoplastic resins used in the laminate according to the present invention include polyolefin such as polyethylene (low density polyethylene, high density polyethylene, linear low density polyethylene, very low density polyethylene, etc.), polypropylene, syndiotactic polypropylene, polybutene and polypentene; polyester such as polyethylene terephthalate and polybutylene terephthalate; polyamide such as nylon 6 and nylon 66; ethylene-ethyl acrylate copolymers, ethylene-tinyl acetate copolymers and polycarbonate. Among these, polyethylene and polypropylene are preferred.

[0029]

In the present invention, various kinds of components such as additives may be incorporated into the alicyclic polymers or the thermoplastic resins as needed. No particular limitation is imposed on the additives so far as they are those generally used in the field of resin industry. Examples thereof include hardening agents, hardening accelerators, hardening aids, fillers, heat stabilizers, weather stabilizers, flame retardants, leveling agents, antistatic agents, slip agents, antiblocking agents, anti-clouding agents, lubricants, dyes,

. 1 . .

5

pigments, natural oils, synthetic oils, waxes, antioxidants, ultraviolet absorbents, light stabilizers, colorants, etc.

These additives may be used in a proper amount within limits not impeding the objects of the present invention.

[0030]

In the present invention, a rubbery polymer may be incorporated into the alicyclic polymers or the thermoplastic resins from the viewpoints of imparting strength or flexibility to the resulting laminate.

10 Examples of the rubbery polymer include diene rubbers such as natural rubber, polybutadiene rubber, polyisoprene rubber, acrylonitrile-butadiene copolymer rubber, styrenebutadiene copolymer rubber, styrene-isoprene copolymer rubber and styrene-butadiene-isoprene copolymer rubber; hydrogenated products of these diene rubbers; saturated 15 polyolefin rubbers, such as ethylene-lpha-olefin copolymers such as ethylene-propylene copolymers, and copolymers of propylene with another lpha-olefin; lpha-olefin-diene copolymer rubbers such as ethylene-propylene-diene copolymers, $\alpha\text{--}$ olefin-diene copolymers, isobutylene-isoplene copolymers 20 and isobutylene-diene copolymers; special rubbers such as urethane rubber, silicone rubber, polyether rubber, acrylic rubber, propylene oxide rubber and ethylene-acrylic rubber; thermoplastic elastomers, such as aromatic vinyl polymers such as styrene-butadiene-styrene block copolymer rubber 25 and styrene-isoprene-styrene block copolymer rubber, and hydrogenated products thereof; urethane-based thermoplastic

elastomers; polyamide-based thermoplastic elastomers; and 1,2-polybutadiene-based thermoplastic elastomers.

[0031]

The rubbery polymers may be used either singly or in

any combination thereof. The blending proportion of the
rubbery polymer is generally at most 100 parts by weight,
preferably at most 70 parts by weight, more preferably at
most 50 parts by weight per 100 parts by weight of the
alicyclic polymer or thermoplastic resin, and the lower

limit thereof is 0 part by weight.

[0032]

15

The laminate according to the present invention may be any laminate so far as it is a laminate obtained by laminating the alicyclic polymer layer, the layer containing the alicyclic polymer and the thermoplastic resin, and the thermoplastic resin layer. These layers may be each provided as either a singly layer or plural layers.

Specific examples of the laminate according to the present invention include a laminate obtained by laminating the thermoplastic resin layer, the layer containing the alicyclic polymer and the thermoplastic resin, and the alicyclic polymer layer in this order; a laminate obtained by laminating the thermoplastic resin layer, the layer containing the alicyclic polymer and the thermoplastic resin, the alicyclic polymer layer, the layer containing the alicyclic polymer layer, the layer containing the alicyclic polymer layer, the layer containing the alicyclic polymer and the thermoplastic resin, and the thermoplastic resin layer in this order; and a laminate

25

obtained by laminating the thermoplastic resin layer, the alicyclic polymer layer, the layer containing the alicyclic polymer and the thermoplastic resin, the alicyclic polymer layer and the thermoplastic resin layer in this order. The layer containing the alicyclic polymer and the thermoplastic resin is interposed between the thermoplastic resin layer and the alicyclic polymer layer, whereby moisture permeability can be reduced, and moreover tearability or cuttability can be enhanced, and separation at an interface between the individual layers can be made hard to occur.

The laminate containing a structure such that the thermoplastic resin layer, alicyclic polymer layer, the layer containing the alicyclic polymer and the

15 thermoplastic resin, alicyclic polymer layer and the thermoplastic resin layer are laminated in this order is most preferable because the laminate can be provided as a laminate having high surface smoothness by holding the layer containing the alicyclic polymer and the

20 thermoplastic resin between 2 alicyclic polymer layers.

[0033]

The thickness of the laminate according to the present invention may be suitably selected as necessary for the end application intended. The thickness of the laminate is of the order of generally 0.5 μ m to 5 mm, preferably 1 μ m to 2 mm, more preferably 5 μ m to 0.5 mm.

In the laminates according to the present invention,

no particular limitation is imposed on the thickness of each of the thermoplastic resin layer, the layer containing the alicyclic polymer and the thermoplastic resin, and the alicyclic polymer layer. However, from the viewpoint of enhancing transparency, the thickness of the thermoplastic resin layer is generally 0.2 to 250 μ m, preferably 0.5 to 100 μ m, the thickness of the alicyclic polymer layer is generally 0.1 to 180 μ m, preferably 0.3 to 70 μ m, and the thickness of the layer containing the alicyclic polymer and the thermoplastic resin is generally 0.07 to 75 μ m, preferably 0.1 to 30 μ m.

[0034]

.

5

10

The thickness ratio [(alicyclic polymer layer)/(thermoplastic resin layer)] of the alicyclic

15 polymer layer to the thermoplastic resin layer is generally 1/99 to 70/30, preferably 5/95 to 55/45. When this thickness ratio is brought close to a proportion of the alicyclic polymer to the thermoplastic resin contained in the layer containing the alicyclic polymer and the

20 thermoplastic resin in terms of a volume ratio, the production efficiency can be enhanced.

Also the thickness of the layer containing the alicyclic polymer and the thermoplastic resin is generally 5 to 50%, preferably 10 to 35% based on the total thickness of the alicyclic polymer layer and the thermoplastic resin layer. When the thickness of the layer containing the alicyclic polymer and the thermoplastic resin is made small,

the tendency is toward improved transparency. When the thickness is made great, the tendency is toward lowered moisture permeability and improved tearability or cuttability.

5 [0035]

10

15

20

25

The laminates according to the present invention are not particularly limited by the production process thereof. For examples, they can be provided by forming the respective layers by a melt forming method or solution casting method and then laminating the layers on each other. The laminates can also be obtained by melt forming the alicyclic polymer, the thermoplastic resin or a mixture of the alicyclic polymer and the thermoplastic resin making up the respective layers.

As a specific example of the solution casting method, may be mentioned a method in which a bar coater, T-die coater, T-die equipped with a bar, doctor knife, Meyer bar, roll coater, die coater or the like is used. A method of coating by means spraying, brushing, rolling, spin coating, dipping or the like may also be adopted. When the desired coating thickness cannot be achieved by one coating, the coating may be conducted repeatedly.

Specific examples of the melt forming method include melt-extruding methods such as a method making use of a T-die and an inflation method, a calendering method, a hot pressing method, and an injection molding method.

[0036]

A preferable production process of the laminate according to the present invention is a process for producing a laminate, which comprises melting and kneading a laminate material comprising at least one alicyclic polymer layer and at least one thermoplastic resin layer to prepare a kneading product, and then co-extruding the kneading product and other polymers or resins.

[0037]

5

The laminate material used in this production process

10 has at least the alicyclic polymer layer and the
thermoplastic resin layer.

Such laminate materials include waste materials produced upon trimming laminates obtained by laminating an alicyclic polymer layer and a thermoplastic resin layer, laminates obtained by laminating an alicyclic polymer layer, a layer containing an alicyclic polymer and a thermoplastic resin, and a thermoplastic resin layer, or the above-described laminates according to the present invention formed by extrusion or the like into standard sizes, and the like.

[0038]

In the present invention, this laminate material is melt and kneaded and then the kneaded product is extruded to form a layer containing the alicyclic polymer and a thermoplastic resin. The other polymers or resins may be extruded to form layers containing each polymers or resins.

[0039]

The extrusion conditions in the melt extrusion are the same as general conditions used for resin materials having almost the same glass transition temperature as the above-described resins. For example, the resin materials are extruded at a resin temperature of about 200 to 300°C through a T-die or the like, and the extruded laminate is taken up by take-up rolls preset to a temperature of about 40 to 100°C to cool the laminate. In order to lessen surface defects such as die lines, it is preferable to construct an extruder in such a manner that portions at which the resin remains are lessened as much as possible. Therefore, it is preferable to use a die free of flaw and the like within the interior or lip thereof as much as possible.

15 [0040]

10

20

In order to laminate the respective layers, the resin materials for forming the respective layers may be coextruded. From the viewpoint of dimensional stability, however, it is preferable to separately extrude the resin materials for forming the respective layers and then laminate the respective layers by calendering, rolling or the like.

[0041]

The laminates according to the present invention may

be used not only in optical materials but in a wide variety

of fields. For example, the laminates in the form of a

film or sheet may be used in packaging films, agricultural

films, conductive films, light diffusing films, electronic circuit boards, insulating sheets, etc. The laminates in the form of a container may be used in chemical containers, drug containers, liquid chemical containers, food

5 containers, drink containers, infusion bags, etc. Besides, they may also be used in light diffusing plates, light guiding plates, liquid crystal substrates, etc.

[0042]

[EXAMPLES]

- The laminate according to the present invention and the production process thereof will hereinafter be described more specifically by the following Examples.

 Evaluations were conducted in accordance with the following respective methods:
- 15 (1) Haze was measured in accordance with ASTM D 1033.
 - (2) Appearance upon stretching:

After stretching a laminate sample, the appearance of the sample was observed and evaluated on a scale of A (good) to D (bad) by degree of clouding, wrinkling and torn line occurred.

(3) Smoothness:

20

The appearance of a laminate sample was visually observed to evaluate it as to smoothness on a scale of A (good) to D (bad).

25 (4) Cuttability:

An aluminum-made cutter attached to an outer case for household food packaging laminate was used to cut a

laminate sample, thereby evaluating it as to easy cuttability at this time on a scale of A (good) to D (bad). [0043]

Referential Example

A ring-opening copolymer of tricyclo[4.3.0.1^{2.5}]dec-3,7-diene with 8-ethyltetracyclo[4.4.0.1^{2,5}.1^{7,10}]dodec-3-ene was hydrogenated, thereby obtaining a hydrogenated ringopening polymer having a weight average molecular weight of 36,000, a molecular weight distribution (Mw/Mn) of 2.1 and a glass transition temperature of 136°C.

Thirty parts by volume of the hydrogenated ringopening polymer and 60 parts by volume of polyethylene
(degree of crystallinity: 54%, melting point: 121°C, melt
flow rate: 2 g/10 min.) were co-extruded by an extruder to

15 obtain a laminated film having a structure that a
hydrogenated ring-opening polymer layer was held by two
polyethylene layers, a thickness ratio of the
polyethylene/the hydrogenated ring-opening polymer/the
polyethylene of 1/1/1 and a total thickness of about 40 µm.

Comparative Example 1

The laminated film obtained in Referential Example was melted and kneaded. Twenty parts by volume of the kneaded product (content of the hydrogenated ring-opening polymer: about 33% by volume) and 80 parts by volume of polyethylene were co-extruded by an extruder to obtain a laminated film having a structure that a hydrogenated ring-

opening polymer layer was held by two polyethylene layers, a thickness ratio of the polyethylene/the kneaded product of the polyethylene and the hydrogenated ring-opening polymer/the polyethylene of 2/1/2 and a total thickness of about 40 μm .

[0045]

Example 1

5

10

15

A kneaded product obtained by melting and kneading the laminated film obtained in Referential Example, the hydrogenated ring-opening polymer and polyethylene were coextruded by an extruder to obtain a laminated film laminated in order of the polyethylene/the kneaded product /the hydrogenated ring-opening polymer/the kneaded product /the polyethylene and having a thickness ratio among the respective layers of 3/1/2/1/3 and a total thickness of about 56 μm .

[0046]

Example 2

A laminated film laminated in order of a polyethylene

20 /a hydrogenated ring-opening polymer/a kneaded product/a
hydrogenated ring-opening polymer layer/a polyethylene
layer and having a thickness ratio among the respective
layers of 3/1/2/1/3 and a total thickness of about 13 μm
was obtained in the same manner as in Example 1 except that

25 the order of lamination and the thickness ratio of the
respective layers were changed.

[0047]

Example 3

A laminated film laminated in order of a polyethylene /a kneaded product/a hydrogenated ring-opening polymer and having a thickness ratio among the respective layers of 2/2/1 and a total thickness of about 24 μm was obtained in the same manner as in Example 1 except that the order of lamination and the thickness ratio of the respective layers were changed.

[0048]

The evaluation results of the laminated films obtained in Examples 1 to 3 and Comparative Example 1 are shown in Table 1.

[0049]

[Table 1]

15

Table 1

	Comp. Ex. 1	Ex. 1	Ex. 2	Ex. 3
Layer structure	PE/BL/PE	PE/BL/COP/ BL/PE	PE/COP/BL/ COP/PE	PE/BL/COP
Thickness ratio	2/1/2	3/1/2/1/3	3/1/2/1/3	2/2/1
Thickness [µm]	40	56	13	24
Haze	35	26	6	12
Appearance upon stretching	D	В	A	В
Surface smoothness	D	С	A	В
Cuttability	D	A	А	A

[0050]

In the table 1, PE represents polyethylene, COP represents hydrogenated ring-opening polymer layer, and BL represents kneaded product of PE and COP.

20 [0051]

[Effects of the Invention]

The laminates or the laminated films according to the present invention are free of clouding, high in transparency and heat resistance, excellent in tearability or cuttability, low in moisture permeability and good in adhesion to others, and thus can be used in a wide variety of use applications such as food packaging films, drug packaging films, agricultural films.

[Document Name] ABSTRACT

[Abstract]

[Object] To provide a laminate which is free of clouding, high in transparency and heat resistance, excellent in tearability or cuttability, low in moisture permeability and good in adhesion to others, and a production process thereof.

[Constitution]

alicyclic polymer layer, at least one layer containing an alicyclic polymer and a thermoplastic resin, and at least one thermoplastic resin layer. To obtain a process for producing a laminate which comprises melting and kneading a laminate material comprising at least one alicyclic polymer layer and at least one thermoplastic resin layer, and then co-extruding the kneaded product and other polymers or resins.

[Selected Figure of Drawings] None